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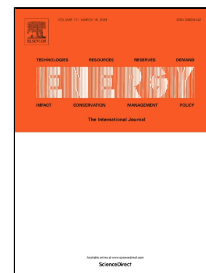
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# Accepted Manuscript

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## ***An experimental study of the drag reducing surfactant for district heating and cooling***

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*Declarations of interest: Beraid DR-IW616 and DR-IW618 were created by AkzoNobel. Beraid DR-IW616 was tested at AkzoNobel testing facility.*

### **Abstract:**

Drag reducing surfactants can be applied to decrease pressure losses in closed circulation pipelines systems like district heating and cooling. This article represents empirical lab scale study of the two drag reducing products developed by AkzoNobel (Sweden) specifically for aqueous solutions for different temperature ranges. Two main issues stopped previous researches of drag reducing agents: heat transfer reduction in shell and tube heat exchanger and environmental concerns. Current shift to heat supply based on renewable energy sources and small-scale biogas CHP plants will no longer require such kind of heat exchangers. It opens opportunities for new surfactants and its application in district heating. This research outlines environmental properties of the two surfactants including its biodegradability rates.

**Keywords:** district heating, drag reducing agent (DRA), surfactant, pressure drop.

### **1. Introduction**

Low concentration of drag reducing agents in turbulent flows of aqueous solutions can cause significant pressure loss reduction. It was first discovered in 1948 by B.A. Toms. He found that the critical Reynolds number of the diluted polymer solution is higher than in water without the additive [1,2]. Sometimes such drag reduction is called “Toms effect”. However, tests that were conducted by Forrest and Grierson already in 1931 showed a reduction in pressure loss in turbulent flow of a wood pulp fiber suspension [3]. All drag reducing agents can be classified into three groups: fibers, polymers and surfactants.

1. The fiber suspensions give significant drag reduction but they are not suitable for district heating because of possible clogging of the system[4].
2. The polymers are friction reducing agents that were extensively studied. The main disadvantage of polymers is that they cannot regenerate after mechanical or thermal degradation in the pipes [5].

3. The surfactants were invented in the 80s. Their main advantage is the ability to reform after destruction what is necessary in closed circuits such as district heating and cooling [6].

First commercial application of drag reducing agents (DRA) namely polymers began in the petrochemical industry. Until nowadays, polymers are added to oil in long transmission pipes to reduce the pumping power. The most famous application is the Trans-Alaska Pipeline system, which is around 800 miles. DRAs are successfully used there to reduce the number of pumping stations [2,3]. Also polymers are supplemented to heated water that surrounds oil to keep it at higher temperature to decrease pressure losses [10,11].

Full scale applications of surfactant solution in district heating were done in Herning, Denmark in the transmission double pipes with diameter of 200 mm and length of 2,8 km [11,12]. Pipes were isolated from the rest of the system with two plate heat exchangers (PHE). Figure 1 shows the results of the study. Supply temperature and return temperature are equal to 80°C and 60-75°C respectively. Significant pressure loss reduction of 70-80% was achieved. Studies were stopped because of heat transfer reduction problem in shell and tube heat exchanger.

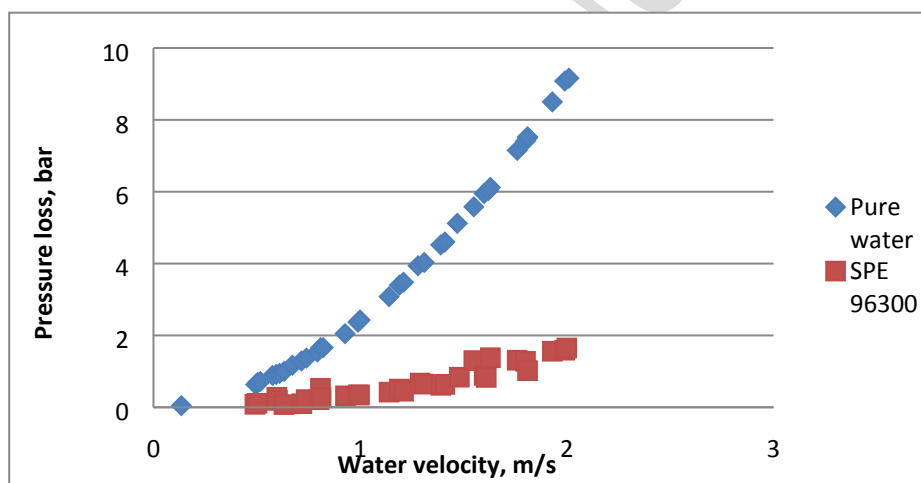


Figure 1: Formation of wormlike micelles

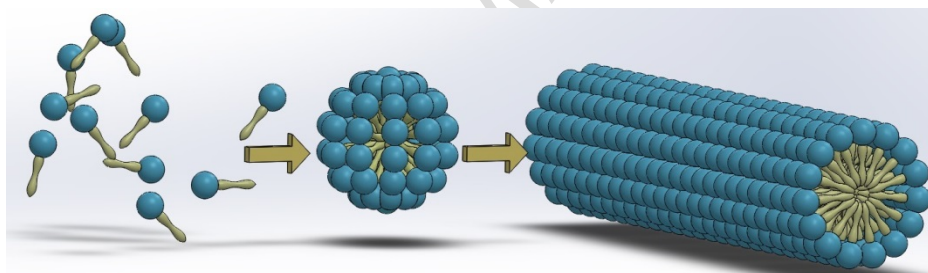
Also similar researches were carried out in Volklingen, Germany and Prague, Czech Republic [18,19]. Another area of application is district cooling where surfactant should be designed for temperature range 5-45°C. Large scale test was done in Japan where surfactant was supplemented to water in air conditioning system of the office building. Energy conservation reached 47% during cooling period [15]. Currently DRA application studies in district heating are focused on finding solutions to such problems as decreased heat transfer in heat exchangers by using different methods of stressing the flow over its path [12,13,14]. These tests also investigate different parameters that can help to find best operational ranges for drag reducers, i.e. critical concentrations and critical temperatures [15,16].

## 2. Theory of drag reduction

Surfactant molecules are amphiphilic compounds, consisting of hydrophobic long tails and hydrophilic heads. There are different types of surfactants depending on the charge of the head-group and they could be either non-ionic, anionic, cationic or zwitterionic [21]. The zwitterionic surfactants have both negative and positive charges in the same molecule. Together with cationic surfactants, positively charged, they are the most useful surfactants as drag reducing agents. The most studied type is the cationic surfactant [7].

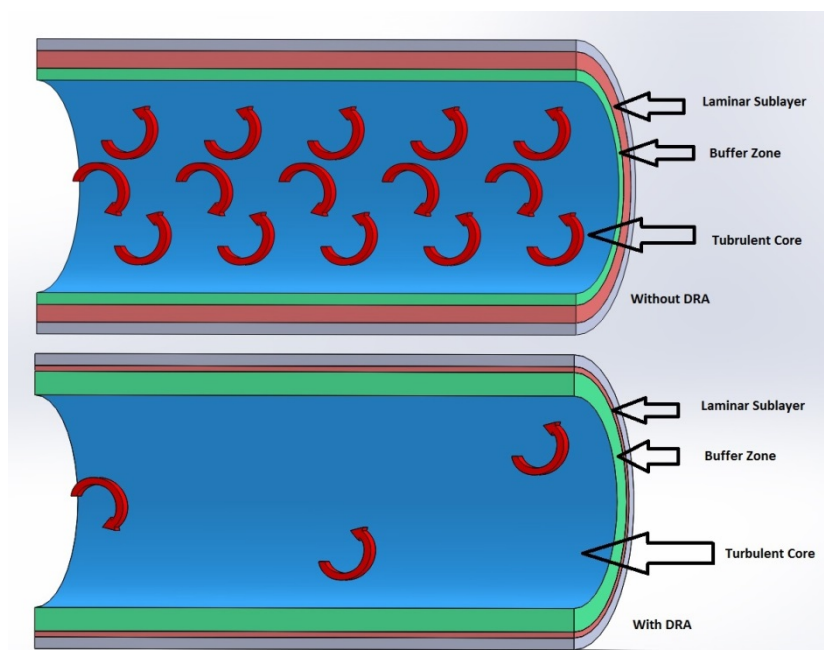
Micellization, the formation of surfactant aggregates, so-called micelles, is a consequence of the driving force to minimize contact between water and hydrophobic chains. Molecules gather in such way that hydrophilic heads form the surface of the micelle and the hydrophobic groups interact and hide in the core of the micelle [22]. The formation of long entangled chains of cylindrical micelles is following the concept of the Critical Packing Parameter (CPP). The most simple version was introduced by Israelachvili in 1976 [23]. It calculates the ratio of cross-sectional area of surfactant tail group to that of the head group. The CPP for cylindrical or worm-like micellar systems is typically  $1/3 - 1/2$ .

The process and different stages of the formation of the worm-like micelles, designed with the packing parameter explained above, are represented in Figure 2. It starts with single molecules of the surfactants in aqueous solution which after reaching a critical concentration gather into the spherical micelles and after reaching a second critical concentration, wormlike micelles or long chained structures are created [7,8].



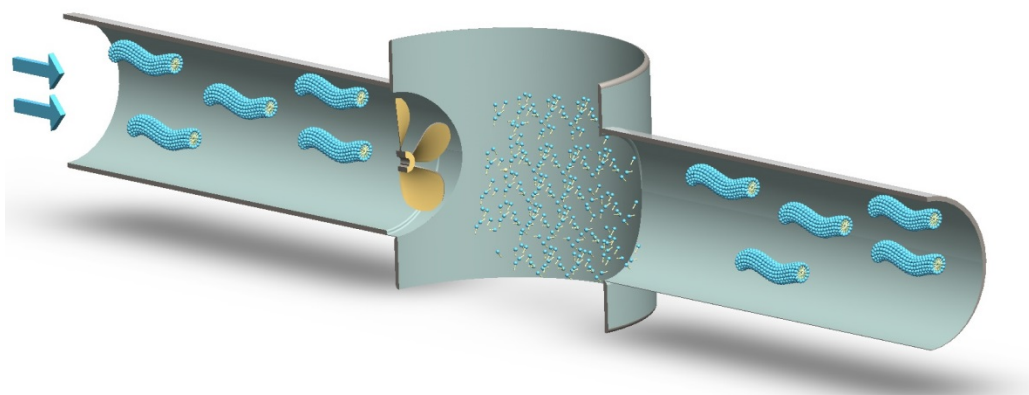
*Figure 2: Formation of wormlike micelles*

As a result, these chains serve as a buffer between the turbulent eddies and consequently decrease turbulence of the flow what leads to lower pressure losses and less pumping energy. Another factor of pressure loss reduction is viscous sublayer that is formed on the wall of the pipes and reduce friction between fluid and pipe. The dampening mechanism of the turbulent bursts are shown in Figure 3.



*Figure 3: Turbulence core change using DRA*

Reformation of torn micelles after crossing the pump is illustrated in Figure 4. Figure 5 shows experimental friction factor asymptotes which were created by Virk for polymers and by Zakin for surfactants.



*Figure 4: Rearrangement of broken micelles*

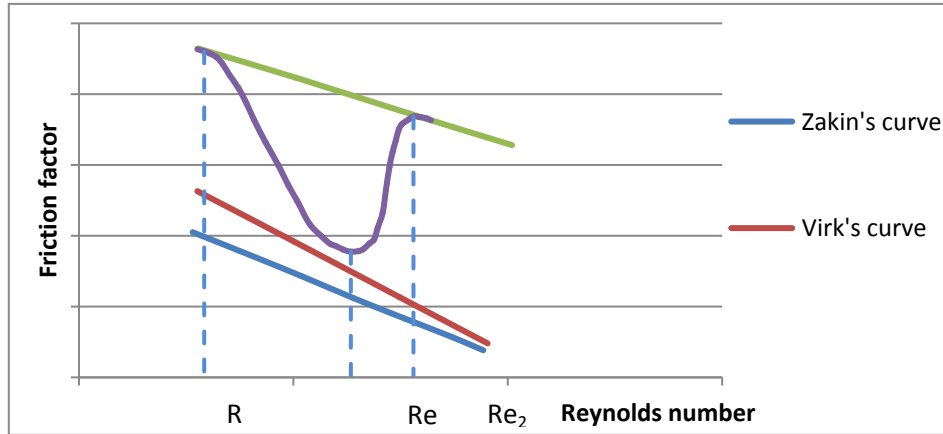


Figure 5: Asymptotes of friction factors for drag reducing agents.  $Re_1$  Reynolds number after which drag reduction is observed;  $Re_c$  critical Reynolds number where micelles structure is torn and drag reduction decreases;  $Re_2$  Reynolds number when drag reduction disappears completely [14]

Virk's asymptote 
$$\frac{1}{\sqrt{f}} = 19 \cdot \lg (Re \cdot \sqrt{f}) - 32.4 \quad (1)$$

Zakin's asymptote 
$$f = 0.315 \cdot Re^{-0.55} \quad (2)$$

$$Re = \frac{\rho \cdot v \cdot d}{\mu} \quad (3)$$

$$DR = \frac{f_0 - f}{f_0} \quad (4)$$

### 3. Methods

#### Working fluids

Beraid DR-IW 616 and 618 are drag reducing agents, developed by AkzoNobel for aqueous solutions. Beraids consist of two surfactants zwitterionic and anionic. Such types combination was first studied by Martin Hellsten and Ian Harwigsson in 1996 [26]. Both products include same type of surfactants with different design to meet different temperature requirements. Beraids were diluted in deionized water with sodium nitrite concentration of 1g/l.

#### Test facility

Tests were carried out independently at AkzoNobel plant in Sweden, where Beraid DR-IW 616 was tested. Beraid DR-IW 618 was investigated at Aalborg University. Experimental setups are shown in the Figure 6 and Figure 7. Both test rigs are similar in main components: centrifugal pump (flow regulation performed by frequency controller), water tank, expansion

tanks and straight sections of pipes, where pressure drop is measured. Setup in Sweden has two loops of 8 mm and 10 mm and three intervals of measuring pressure drop at each loop. First pressure tap located 1 meter from 180° turn. Two variable area flowmeters were used for measuring water flow with deviation of measurement of  $\pm 5\%$ . Pressure measured by differential pressure gauge with uncertainty of  $\pm 2\%$ . System of valves was used to measure pressure drop of different pipe sections with one sensor.

Beraid DR-IW 618 was tested in  $\frac{3}{4}$ " pipeline at Aalborg University. Ultrasonic flowmeter was used to measure the flow. Deviation was  $\pm 1,5\%$  of measuring value. Pressure drop was measured by piezoresistive differential pressure sensor where the uncertainty was  $\pm 2\%$ . National Instrument data acquisition system was used to collect data from sensors.

Validation of the test rigs was done by comparing theoretical pressure loss and experimental results of pure water. Deviation of maximum 5% was achieved.

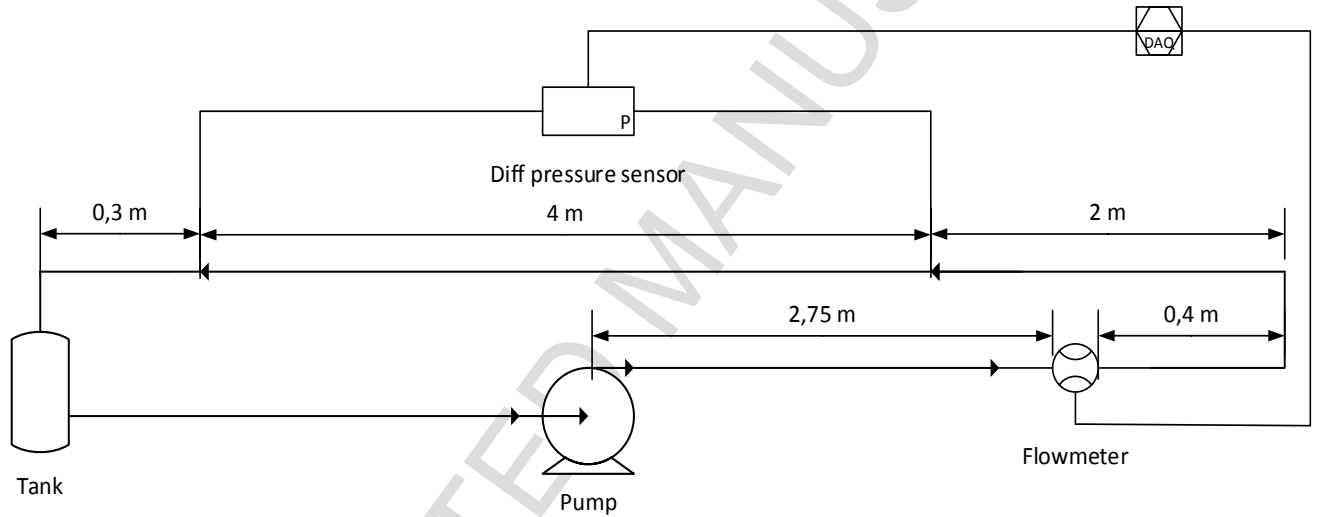


Figure 6: Test setup at Aalborg University



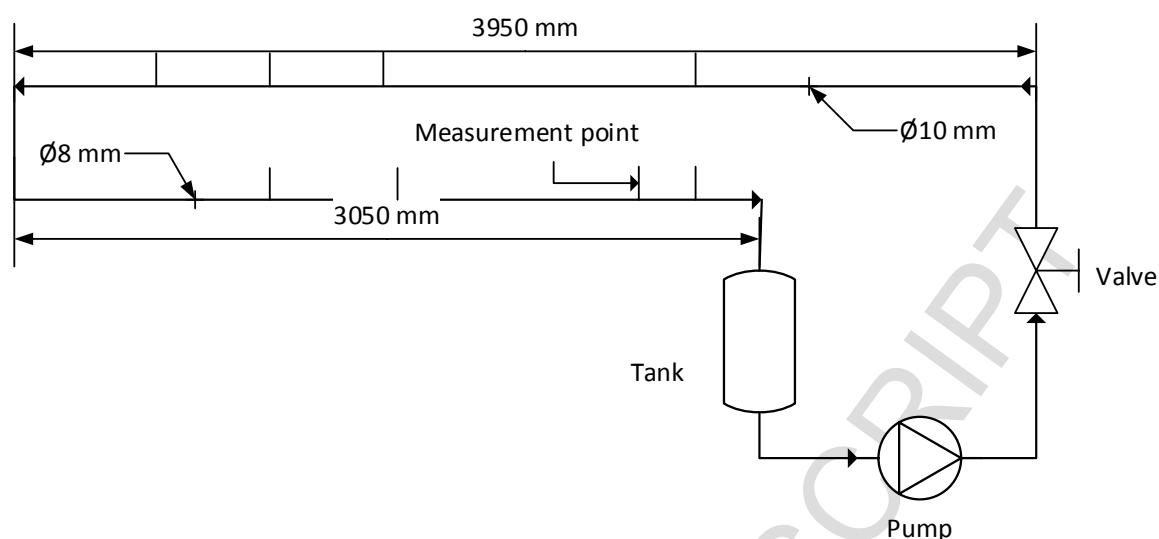


Figure 7: Test setup at Akzo-Nobel

#### 4. Results

Figures 8-9 illustrate Moody's friction factor of Beraid DR-IW 616 solution at 25°C and 40°C respectively. Drag reduction from 60% to 80% is achieved for a flow from 0,5 m/s to 3,5 m/s with concentration of 1250 ppm at 25°C. Pressure loss decrease of 70% - from 0,25 m/s to 2,25 m/s at 40°C 1000 ppm. Lambda O1, O2, O3 represent drag reduction at different sections of pipes.

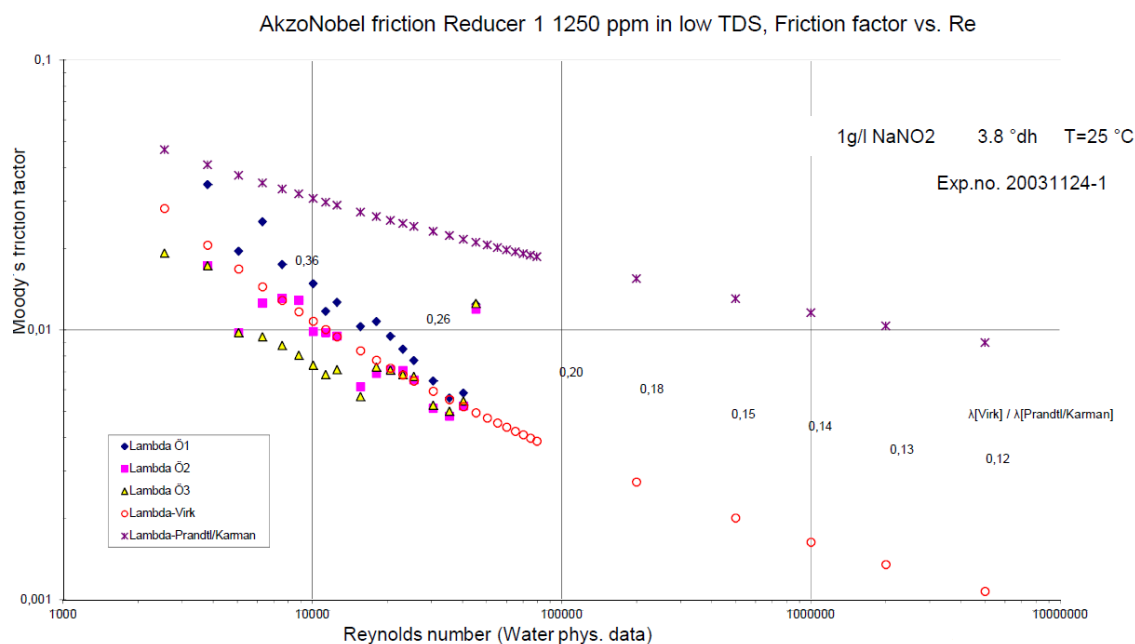


Figure 8: Beraid DR-IW 616 at 25°C

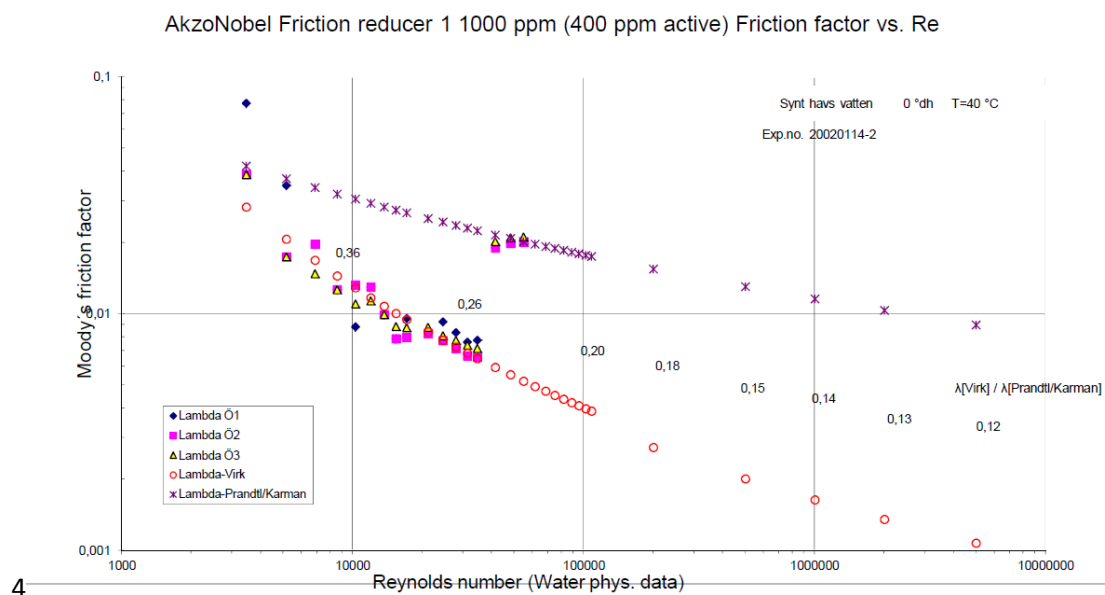


Figure 9: Beraid DR-IW 616 at 40°C

Figure 10 displays comparison of Fanning friction factor between pure water, Zakin's asymptote and surfactant solution of Beraid DR-IW 618 at different temperatures. Drag reduction disappears at certain flow velocity when critical shear stress is reached and structures are torn and cannot form new micelles. Beraid DR-IW 618 did not show any drag reduction at 60°C. Obviously, effect disappears between 55°C and 60°C. This was also proven by beaker test (developed by Akzo Nobel), where absence of vortex formation of 0,1% surfactant solution in 40 ml beaker, during stirring, indicated loss of a drag reduction properties.

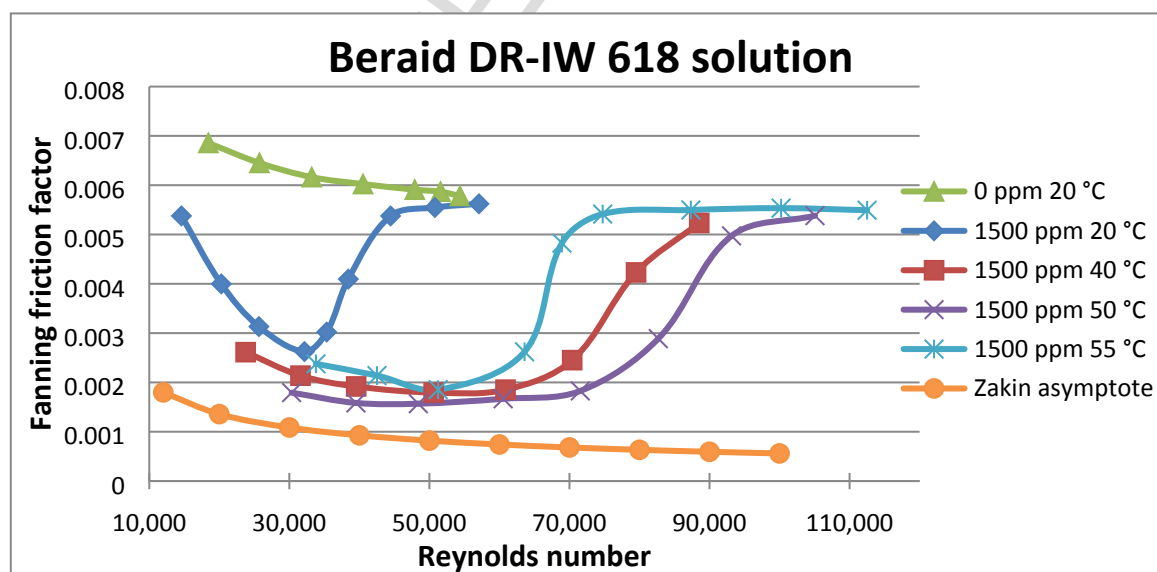


Figure 10: Friction factors of Beraid solution at different temperatures

Pressure loss saving of Beraid DR-IW 618 solution are presented in the Figure 11. Higher effects are reached at temperature range from 40°C to 55°C where drag reduction 60-74% was obtained.

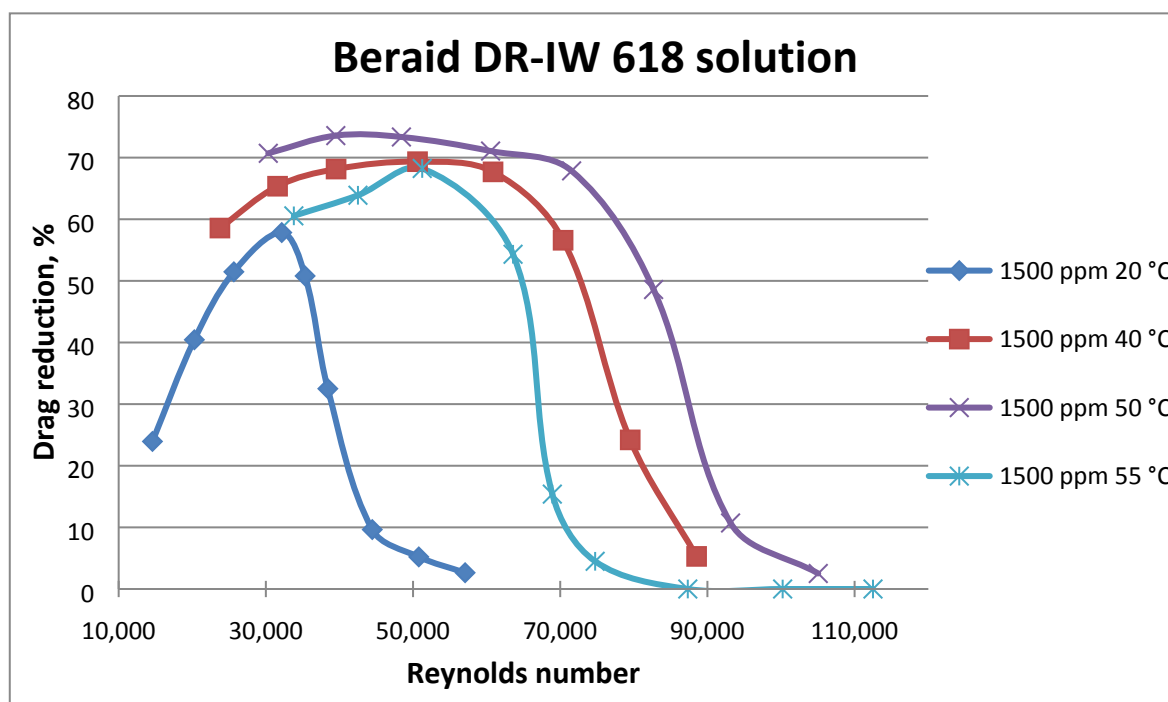


Figure 11: Drag reduction of Beraid solution at different temperatures

According to the conducted experiments, Beraid DR-IW 616 has a higher maximum drag reduction of 70-80% when Beraid DR-IW 618 has 60-75%. These studies were carried out in pipes of different diameter. Lower effect of Beraid DR-IW 618 partially can be explained by difference in pipe size, which can cause change in pressure loss reduction [27].

## 5. Environmental aspects

The environmental impact of surfactants is often divided into toxicity, biodegradation and bioaccumulation. The type of surfactant, its surface activity and specific adsorption determines the effect in the environment. The most important and noticeable action of surfactants is the adsorption to biological surfaces of aquatic living organisms causing toxic effects to algae, daphnia and fish. Another important factor in minimizing the effect of surfactants is the ability to degrade in the environment. Biodegradation is dependent on the structure of the hydrophobic and hydrophilic moiety of the surfactant which has to be designed so it can be cleaved and biodegraded under aerobic and preferably also under anaerobic conditions.

In the early development of DRA cationic surfactants were the dominating type in the research for commercial drag reducing surfactants. Cationic additives together with anionic counter ion like salicylate showed good ability to form thread-like micelles and gave a good degree of drag-reduction. However, the cationic surfactants carrying a permanent cationic

charge are toxic to aquatic organisms due to the high degree of adsorption onto the membranes and tissues of water living organisms. High toxicity in combination with limited biodegradation confined the use of cationic surfactants in district heating and cooling water applications.

The zwitterionic acylamidopropylbetaine surfactant, which was used in Beraid DR-IW 616 and in Beraid DR-IW 618, has improved environmental properties compared to cationic surfactants. Zwitterionic surfactants like the acylamidopropyl betaine is a very common ingredient in liquid soaps, hair shampoos and shower gels. They show less toxicity to aquatic life compared to cationic surfactants and they have high degree of biodegradation. The zwitterionic component in the Beraid DR-IW 616 is easily biodegradable in fresh water, reaching 93% in 28 days according to OECD method 301 D. It was also found to be readily biodegradable in a seawater, 70 % was biodegraded in 28 days according to closed bottle test OECD 306. The anionic surfactant in Beraid DR-IW 616 is a fatty alcohol sulphate. This surfactant belongs to the oldest surfactant type, which has also found widespread use in liquid detergents, hair shampoos and shower creams. Alkyl sulphates are easily biodegradable in both seawater and fresh water. They are also degraded under anaerobic conditions. The biodegradation of the zwitterionic surfactant of Beraid DR-IW 618 is expected to be similar to Beraid DR-IW 616, meaning readily biodegradable in fresh and seawater.

The favourable ecotoxicology properties of the surfactant components in Beraid drag reducers make them useful for district heating and cooling applications. In the Herning trials in the late 90's similar surfactant components were used and approved by Danish authorities.

## 6. Discussion

Achieved results, which were presented in previous chapters, show huge potential of possible application of Beraid products. Beraid DR-IW 618 demonstrated drag reduction properties from 20°C to 55°C, which is suitable for low-temperature district heating where supply temperature is 50°C and return is 25-30°C. Beraid DR-IW 616 should be tested at lower temperatures to prove expected abilities of drag reduction appropriate for district cooling with operating temperature range 5-45°C.

Possible ways of application of drag reducing surfactants:

- Direct pump energy saving.
- Increase of a flowrate remaining pumping energy on previous level. Low-temperature district heating is a key area for such application where maintaining heat supply on the level of 3<sup>rd</sup> generation district heating could demand higher flowrates.
- Attachment of more customers without additional pumping stations and rearranging district heating grid and heat source.
- Smaller pipe diameters for newly designed networks.

Disadvantages that stopped implementation of surfactant to real district heating and cooling networks:

- Environmental concerns stopped further development during some of previous studies. High biodegradability of Beraid products should increase chances of acceptance of the product by authorities.
- Heat transfer reduction in heat exchangers. In order to fulfil Denmark's goal of heat supply based on renewable energy sources, more heat pumps, electric boilers, thermal storages and biogas CHP plants will be used. Also increasing insulation and reconstruction of buildings will decrease demand of heat energy [26, 27]. This all will lead to lower number of large scale CHP plants in Denmark where mainly shell and tube heat exchangers are used. Such type showed highest heat transfer reduction rates, which was one of the main reasons to turn down further implementation of surfactants. Though DRAs performed significantly better in plate heat exchangers. Beraid products are expected to have even smaller heat transfer reduction in PHE because of smaller critical shear stress, which will help to stop drag reduction and eventually heat transfer reduction in plate heat exchangers.

## 7. Conclusion

Drag reduction properties of two commercially available surfactants, produced by AkcoNobel (Sweden), were investigated. Maximum drag reduction of 60-75% for Beraid DR-IW 618 and 70-80% for Beraid DR-IW 616 were achieved after broad testing for different temperatures and velocities. Operating temperature range for Beraid DR-IW 618 is from 20°C to 55°C and expected range for Beraid 616 is from 5°C to 45°C. The different surfactant systems always cover a specific temperature range and if higher temperature requirements are of interest, the surfactant formulation can be tuned to accomplish temperatures up to 120°C.

Both Beraids are fast biodegradable aerobically and anaerobically and have high potential for acceptance by environmental authorities. Issue of heat transfer reduction in shell and tube heat exchanger may disappear because of shift to smaller CHP plants with alternative fuels, solar plants, heat pumps, where plate heat exchanger can be used. This gives an opportunity to achieve critical shear stress easier to break the bonds between surfactants and get water-like behaviour of the solution.

Future studies should focus on heat transfer reduction problem, possible environmental impact and economic analysis including additional costs for heat transfer enhancement measures.

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**Highlights**

- Drag reducing agents can reduce pressure losses in pipes. They were found in 1948
- Process of formation from spherical to rod like micelles
- Lab scale study of two commercial surfactants. Drag reduction of 60-80% was reached
- Tested surfactants showed high biodegradability in fresh and seawater
- Future studies should focus on heat transfer reduction problems, environmental impact